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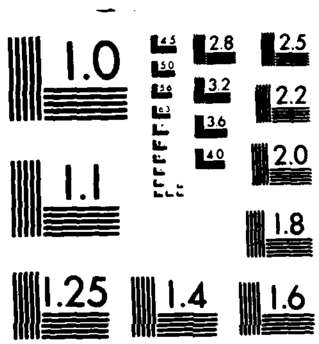

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F-Atom Depletion in NF_3 Combustor Laser Flows

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This report has been reviewed by the Public Affairs Office (PAS) and is releasable to the National Technical Information Service (NTIS). At NTIS, it will be available to the general public, including foreign nations.

This technical report has been reviewed and is approved for publication. Publication of this report does not constitute Air Force approval of the report's findings or conclusions. It is published only for the exchange and stimulation of ideas.

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20. ABSTRACT (Continue on reverse side if necessary and identify by block number)

Chemical lasers employing NF_3 combustors operating at temperatures below 1900°K are subject to F atom depletion because of NF_x (x=1,2) radical chemistry. Experiments using arc-heated NF_3-He mixtures have shown that the F atom flow from stagnation temperatures of 1000 to 1600°K is significantly reduced while considerable NF chemiluminescence is observed. A method for obtaining F atom flow from low temperature combustors is described.

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CONTENTS

FIGURES

I. BACKGROUND

High power HF/DF chemical lasers operate by the reaction of F atoms flowing from a supersonic nozzle. The loss of F atoms by recombination on the nozzle walls and the resulting loss of performance has led to sophisticated nozzle designs that mitigate the problem. A diagnostic for measuring F_2 in nozzle flows was developed to assess the extent of F atom recombination in some of the more advanced designs. It was determined when the instrument was tested on a small, water-cooled copper nozzle attached to an NF_3 combustor that there was virtually no F_2 in the flow, an implication that wall recombination was being masked by other more significant chemical processes going on in the flow. At the same time, the presence of a strong yellow emission from the flow suggested that NF_x radical chemistry was involved. Therefore, it is possible that loss in performance of chemical lasers using NF_3 combustors may also be caused by F atom depletion in the very rapid reactions involving NF_x radicals. Additional tests were made with arc-heated NF_3 flowing through the nozzle to determine conditions making these reactions significant in limiting F atom flow for lasing.

II. EXPERIMENTAL SETUP

A sensitive F_2 diagnostic has been developed[1] to evaluate laser nozzle performance in terms of F atom recombination on the nozzle walls. A small diameter beam of 325 nm radiation is projected through the flow field and is moved so that it passes through the boundary layer coming from the nozzle wall. Any F_2 in the path of the beam will result in some absorption of the beam; with proper calibration, one can measure F_2 concentration with a $\Delta I/I_o$ sensitivity of 3×10^{-5}. The technique has been demonstrated on arc-heated F_2 and SF_6 flows in a water-cooled copper nozzle[1] and on combustion-heated F_2 in a gas-cooled nickel nozzle.[2]

A small calibration facility was set up in the Aerophysics Laboratory to test a variety of nozzle designs with the F_2 diagnostic apparatus. Since most of the nozzles of interest were being operated on NF_3 combustors, a small NF_3 combustor was installed in place of the arc-heated lasers common in the laboratory. A schematic of the system is shown in Fig. 1 and a photograph of the hardware is shown in Fig. 2. All parts are water-cooled. The combustor is 2 by 1-1/2 by 2 inches. The injector was copied from an early TRW design using nine triplets in the pattern, fuel-oxidizer-fuel. The nozzle for the initial checkout tests was a water-cooled copper assembly of modular design, arranged so that each nozzle blade could be removed so its configuration could be changed. The particular configuration tested first had been probed extensively to evaluate its flow characteristics. For most tests, fuel would not be injected at the exit of the nozzle; therefore, fuel was injected through a probe downstream to react the fluorine in the flow before it reached the oil in the pumps of the exhaust system. The combustor was ignited with a repetitively pulsed spark as the fuel flow was increased. A starting sequence is shown in Fig. 3.

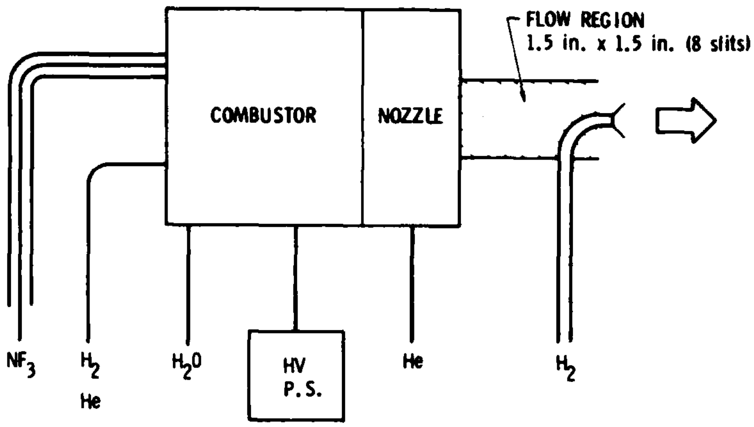

Fig. 1. Facility schematic

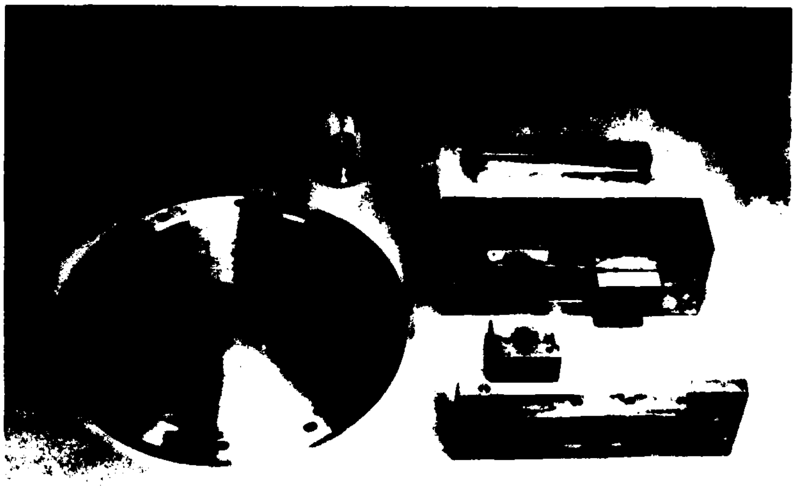

Fig. 2. Combustor and nozzle parts

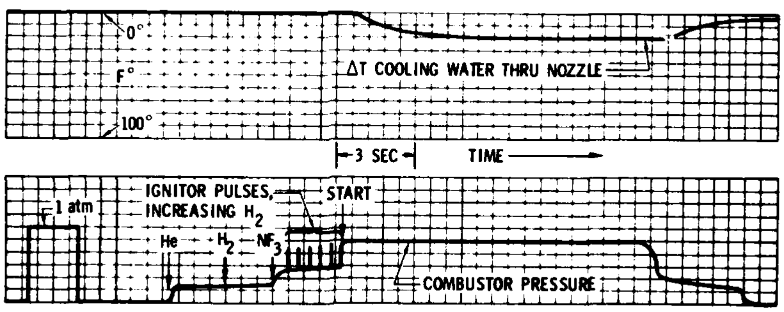

Fig. 3. Combustor starting sequence, $R = .85$, $\beta = 10$, $T_o = 950°K$

III. RESULTS

It was evident from the initial tests that heat losses in the small, water-cooled combustor would preclude operation at the nozzle stagnation temperatures required to simulate laser operation. The run shown in Fig. 3 achieved a temperature of only 950°K. NEST calculations were made for combustion with no heat loss (Fig. 4); the temperature for these operating conditions in an adiabatic combustor would have been 1300°K. Therefore, a 0.055-inch thick alumina liner was retrofitted in the combustor to reduce heat losses.

During these and subsequent tests, strong yellow emission was observed in the flow coming from the nozzle (Fig. 5). The emission had never been observed before in arc-heated F_2 or SF_6 flows. Other work in the laboratory involving NF_x radical chemistry had shown, however, that partial pyrolysis of NF_3 did give characteristic yellow emission generally attributed to N_2 First Positive bands. Furthermore, with the addition of hydrogen, emission in the green at 5288 Å (NF b-X band) could be observed. A green filter was then used on the combustor flow (T_o = 1350°K) and emissions were observed in the region around the probe that injects hydrogen downstream.

When the F_2 diagnostic was used to evaluate F atom recombination on the walls of the nozzle, no F_2 was measured. Previously, water-cooled copper nozzles had always shown significant F_2 in the boundary layers where the ratio of F_2 to the total fluorine flow was 0.50. The probe beam was reoriented to pass through the flow perpendicular to nozzle slits; still, there was no measurable F_2.

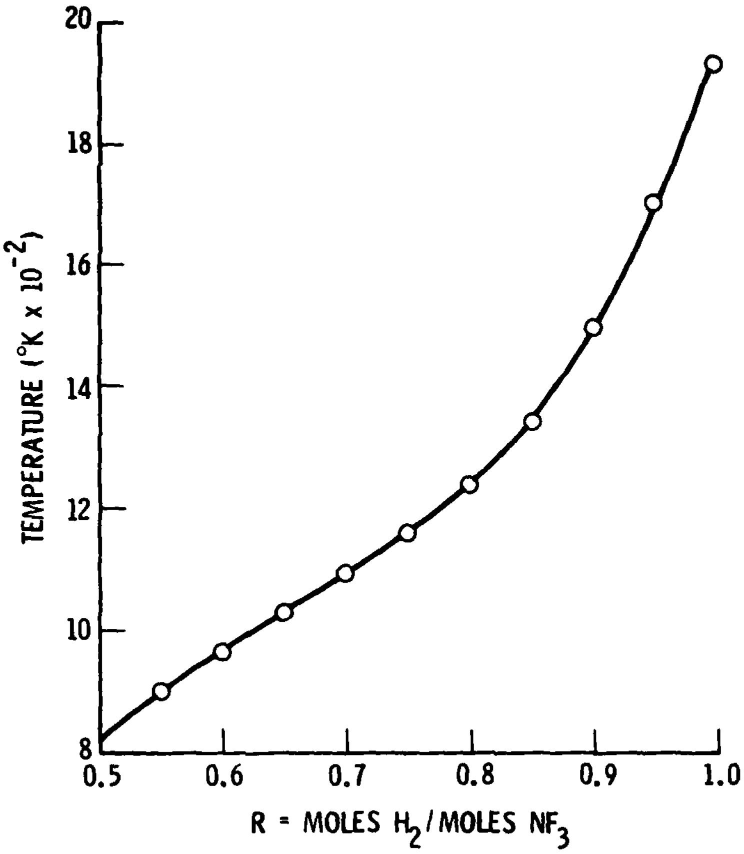

Fig. 4. Flame temperature for NF_3-H_2 combustion in He, no heat loss, p_o = 1 atm, β = 10

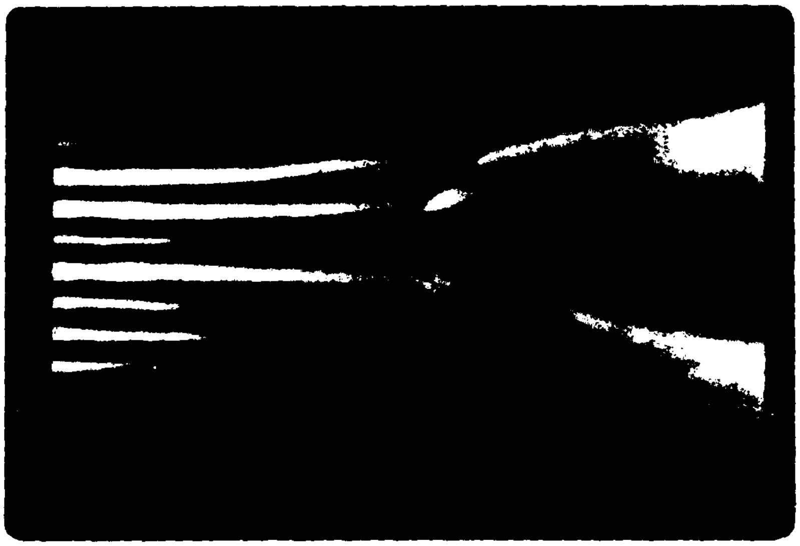

Fig. 5. Yellow glow in nozzle flow from NF_3 combustor, $T_o = 1350°K$

A laser cavity was installed and D_2 was injected through the nozzle. The power measured using a calorimeter[3] was 325 watts. According to standard laser performance criteria, the F atom flow rate required to give this power would be 0.0244 moles per second. If all the NF_3 were completely reacted in the combustor according to the reaction:

$$0.15\ He + 0.047\ NF_3 + 0.040\ H_2 \rightarrow 0.15\ He + 0.0235\ N_2 + 0.080\ HF + 0.061\ F$$

the F atom flow rate would have been 0.061 moles per second. The F_2 diagnostic has about 25 times the sensitivity required to measure the F_2 for these F atom flows given a nozzle wall recombination factor of 0.50.

All these results implied that the NF_x radical chemistry associated with this low temperature NF_3 combustor was significantly reducing the F atom (and F_2) flow coming from it. The nozzle was mounted on an arc heater so that the phenomenon could be explored more easily and so that variations in stagnation temperature could be made in the presence of large heat losses and without changes in the gas mixtures. Results of runs made with stagnation temperatures from 1000 to 2550°K are summarized in Table I. Yellow or green chemiluminescence was evident in the lower temperature runs where no F_2 absorption was present. At the higher temperatures, emission was minimal and the F_2/fluorine ratio of 0.50 was measured.

TABLE I. Summary of fractional number of F atoms recombined in various HF/DF chemical laser nozzle flows.*

Nozzle	Heating	Gases	Temp (°K)	F Recombined
Cooled copper 36 slit	Arc	F_2, SF_6	1400 to 2100	0.50
Hot nickel CL XI B	Combustor	F_2	1000 to 1600	0.10 to 0.20
Cooled copper 8 slit	Combustor	NF_3	1350	<0.01
Cooled copper 8 slit	Arc	NF_3	1000, 1300, 1600	<0.01
			1900, 2550	0.50

*Measured with F_2 absorption diagnostic.

IV. DISCUSSION

The chemical reactions that apply to the NF_3 system are given in Table II. It will be noted that the reactions shown for the high temperature combustor are those used for chemical lasers in which the NF_3 is completely dissociated into its components, N_2 and F (atoms). This appears to be valid for stagnation temperatures above 1900°K in efficient combustors. Below 1900°K the reaction will be incomplete and NF_x radicals will be formed. The equations for the pyrolysis of NF_3 in the arc heater are analogous and do not involve the H_2 and HF.

In the low temperature cases, the presence of NF_x radicals is accompanied by strong emission in the yellow and, with the addition of hydrogen, in the green. It will be noted that the yellow glow of the N_2 transition comes from the reactions involving N and NF, which are components of dissociated NF_3. On the other hand, the green emission of NF comes from the reaction[4] of NF_2 and H and a subsequent reaction with HF(v=2) making HF(v=0), which degrades laser performance. The NF_x radicals are also scavengers of F atoms with recombination rate coefficients (k) three orders of magnitude greater than that for the F atom. It is not surprising, then, that no F_2 was present in the low temperature flow, even with water-cooled copper nozzle walls.

In view of these factors, it is possible that chemical lasers using NF_3 combustors, even equipped with low heat loss components and sophisticated nozzle designs, have not been optimized for best performance. Since efficient use of NF_3 as an F atom carrier requires operation of the combustor at high temperature, the lasable fluorine is diminished. If the diluent in the com-

TABLE II. Chemical Equations for NF_3 System

Combustion:

(Low T)	$NF_3 \xrightarrow{\Delta} NF_2 + F$	
	$F + H_2 \rightarrow HF + H$	
	$H + NF_2 \rightarrow HF + NF$	
	$NF_2 + F \xrightarrow{M} NF_3$	$(k \sim 10^{17})$
	$NF + F \xrightarrow{M} NF_2$	$(k \sim 10^{17})$
	$F + F \xrightarrow{M} F_2$	$(k \sim 10^{14})$
(High T)	$NF_3 \xrightarrow{\Delta\Delta} \frac{1}{2} N_2 + 3F$	
	$F + H_2 \rightarrow HF + H$	$k \sim \left[\frac{1}{sec}\left(\frac{cm^3}{mole}\right)^2\right]$
	$F + F \xrightarrow{M} F_2$	

Arc Heated:

(Low T)	$NF_3 \xrightarrow{\Delta} NF_2 + F$
	$NF_2 + F \xrightarrow{M} NF_3$
	$F + F \xrightarrow{M} F_2$
(High T)	$NF_3 \xrightarrow{\Delta\Delta} \frac{1}{2} N_2 + 3F$
	$F + F \xrightarrow{M} F_2$

Spectroscopy:

$$N(^2D) + NF(a^1\Delta) \rightarrow N_2(B^3\Pi_g) + F$$

$N_2(B^3\Pi_g) \rightarrow N_2(A^3\Sigma^-) + h\nu$ (5500-8800Å) yellow glow

$$H_2 + F \rightarrow HF(2) + H$$

$$H + NF_2 \rightarrow NF(a^1\Delta) + HF(0)$$

$$NF(a^1\Delta) + HF(2) \rightarrow NF(b^1\Sigma^+) + HF(0)$$

$NF(b^1\Sigma^+) \rightarrow NF(X^3\Sigma^-) + h\nu$ (5288Å) green glow bustor is injected after the NF_3 has been completely dissociated, the consumption of fluorine to achieve a given stagnation temperature can be minimized.

We propose a concept that circumvents the problem with low T_o combustors. The situation is shown graphically in Fig. 6 for a D_2/NF_3 combustor. With the addition of diluent downstream, the reaction of D_2 and NF_3 goes to a high temperature, thus ensuring that the NF_3 is completely dissociated. Then the diluent is mixed in to achieve the correct mixture at the required temperature. With this modification, it is conceivable that HF laser performance using NF_3 could optimize at lower combustor temperatures since F atoms available for lasing would be comparable to that obtained with a laser using F_2.

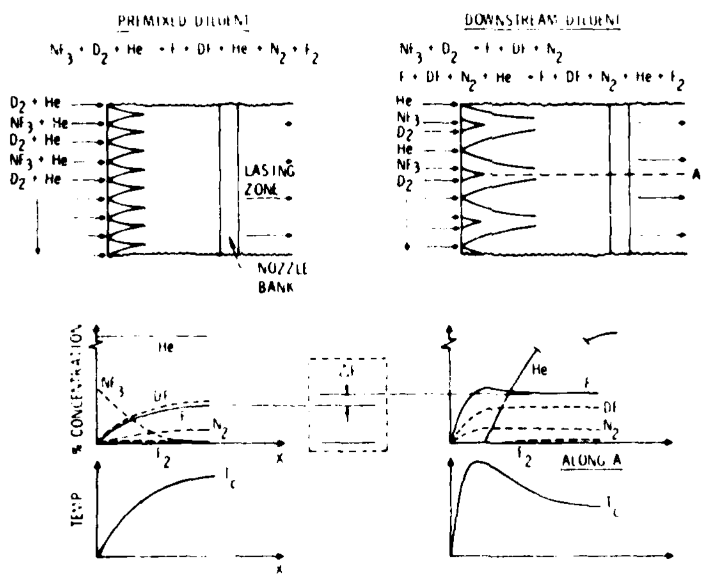

Fig. 6. Downstream dilution concept for D_2/NF_3 combustor

V. CONCLUSION

It was determined during tests of a low temperature NF_3 combustor that the F atom flow was significantly reduced and that there was strong yellow chemiluminescence in the flow. These effects are attributed to the presence of NF_x radicals in the incompletely dissociated NF_3 that scavenge F atoms and give strong spectrographic signatures. Pyrolysis of NF_3 in an arc heater indicated that stagnation temperatures of 1900°K or higher are required to dissociate the NF_3 completely and avoid the losses in laser performance from NF_x chemistry. It is proposed that reacting undiluted NF_3 with an undiluted fuel and adding the diluent downstream in the combustor would result in complete dissociation of the NF_3 at a lower nozzle stagnation temperature with more fluorine available for lasing. Chemical lasers using NF_3 combustors should then perform more like those operating with F_2 combustors.

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